

REMARKS/ARGUMENTS

In view of the amendments to the Specification above and the remarks and arguments below, Applicant believes the pending application is in condition for allowance.

I. Status of the Claims

Claims 1 and 2 are pending and are presented herein as a courtesy to the Examiner. No amendment to the claims is made.

II. Status of the Specification

This application is a U.S. national stage application of International Patent Application No. PCT/JP04/019132, which was filed in Japanese. The PCT application was published as International Publication No. WO 2005/058806 A1.

The present Specification is amended to correct the errors made, without intent to deceive, in preparing an English translation of the PCT application. Attached as Exhibit A are the pages of the WO publication that contain the paragraphs whose English translation is being amended. Exhibit B is a certificate that attests to the accuracy of the translation.

Accordingly, Applicant respectfully submits that no new matter is introduced by the amendments made to the Specification.

III. Perfection of Foreign Priority

Applicant appreciatively thanks the Examiner for the acknowledgment of a claim made in this application for foreign priority from Japanese Patent Application No. 2003-417611 ("JP '611"), filed on December 16, 2003, and for the acknowledgment of receipt of a certified copy of JP '611.

Attached as Exhibit C is a certified English translation JP '611. Exhibit D is a certificate that attests to the accuracy of the translation.

Accordingly, Applicant respectfully submits that the present application is entitled to the benefit of foreign priority from JP '611.

IV. Claim Rejections under 35 U.S.C. § 102(a)

Claims 1 and 2 are rejected under 35 U.S.C. § 102(a) as anticipated by JP 2004-018500 to Osawa (“Osawa”). The Examiner states that Osawa discloses all the elements recited in the claims. Applicant respectfully traverses the rejections.

In view of the remarks presented in the previous section, the present application is entitled to a priority date of December 16, 2003, the filing date of JP ‘611. Osawa was published on January 22, 2004. Since Applicant’s priority date predates Osawa’s publication date, Osawa is removed as prior art.

Accordingly, Applicant respectfully requests that the rejections of claims 1 and 2 on this basis be withdrawn.

V. Claim Rejections under 35 U.S.C. § 103(a)

Claims 1 and 2 are rejected under 35 U.S.C. § 103(a) as unpatentable over CH 680292 A to Chuck et al. (“Chuck”) in view of JP 2003-040854 to Nakamura et al. (“Nakamura”). The Examiner contends that Chuck in combination with Nakamura renders the claims obvious. Applicant respectfully traverses the rejections.

In the present invention as recited in claims 1 and 2, by using sulfur trioxide and cyanogen chloride which are diluted with chlorosulfonyl isocyanate or a solution containing chlorosulfonyl isocyanate, chlorosulfonyl isocyanate can be obtained selectively. Comparison of Examples 1 and 2 on the one hand and Comparative Example on the other shows that only Examples 1 and 2, in which sulfur trioxide and cyanogen chloride diluted with chlorosulfonyl isocyanate or a solution containing chlorosulfonyl isocyanate are used, provide a high yield of chlorosulfonyl isocyanate.

The Examiner states that while “Chuck does not specifically disclose sulfur trioxide and cyanogen chloride respectively diluted with the chlorosulfonyl isocyanate or the solution including

chlorosulfonyl isocyanate, . . . Nakamura teaches that the presence of chlorosulfonyl isocyanate in the reaction mixture enhances chlorosulfonyl isocyanate selectivity (Nakamura, 0011).”¹

However, paragraph [0011] of Nakamura merely states that cyanogen chloride and sulfuric anhydride are mixed and condensed to obtain a condensation reaction solution containing chlorosulfonyl isocyanate, and that the condensation reaction temperature or the reaction rate of cyanogen chloride and sulfuric anhydride can be controlled in order to obtain chlorosulfonyl isocyanate with good selectivity. Nakamura’s paragraph [0011] does not disclose that chlorosulfonyl isocyanate is included in the original reaction solvent, nor does it disclose that this enhances the selectivity of producing chlorosulfonyl isocyanate. Attached as Exhibit E is a certified English translation of Nakamura’s paragraph [0011]. Exhibit F is a certificate that attests to the accuracy of the translation.

Accordingly, Chuck and Nakamura, either alone or in combination, do not disclose all the elements recited in claims 1 and 2. At least for this reason, Chuck in view of Nakamura does not render the claims obvious. Applicant respectfully requests that the rejections of claims 1 and 2 on this basis be withdrawn.

VI. Common Ownership of Subject Matter

Applicant states that the subject matter of claims 1 and 2 was commonly owned by the named joint inventors at the time the invention covered therein was made.

¹ Office Action mailed February 21, 2008, page 5, lines 10-15.

Docket No.: 20241/0207048-US0

In view of the foregoing, it is believed that claims 1 and 2 are in immediate condition for allowance and it is respectfully requested that the application be reconsidered and that all pending claims be allowed and the case passed to issue.

Dated: May 19, 2008

By Laini R. Hutton

Attorneys/Agents For Applicant

Exhibit A

囲でクロロスルホニルイソシアナートを反応溶媒として使用する場合には冷却設備が不要であり、クロロピロスルホイソシアナートを含む副生成物の熱分解処理を同時に行うことができる。

本発明における原料供給時間は、凝縮器の能力範囲内であれば特に限定されない。回分式の場合は、通常0.25～2.5時間である。連続式の場合は、設備の処理能力に応じた原料供給量で反応させればよい。

本発明の製造方法においては、副生成物の分解を完全に行い、収率よく目的とするクロロスルホニルイソシアナートを製造するために十分な反応時間を設けるのが好ましい。反応時間は、原料の消費および副生成物の分解が十分であればよく、通常は0.5～1.5時間、好ましくは5～10時間である。

(4) 後処理操作

反応終了後、反応液を精製して、目的とするクロロスルホニルイソシアナートを単離することができる。反応液の精製は、通常、反応完結後の反応液を常圧下あるいは減圧下において、通常の蒸留塔を用いて蒸留することにより行うことができる。蒸留の条件、例えば、圧力、温度、段数、回数等は特に限定されない。なお、連続式の場合には、前記反応完結槽から連続的に反応液を取り出し、連続的に蒸留することにより、目的とするクロロスルホニルイソシアナートを単離することができる。

以下、実施例および比較例によって本発明をさらに詳細に説明するが、本発明はこれに限定されるものではない。

(実施例1)

凝縮器を取り付けた容量100mlの四つ口フラスコにクロロスルホニルイソシアナート14.3g(0.1モル)を量り取り、攪拌しながら還流温度まで昇温した。この液中に、還流下、三酸化硫黄40.4g(0.5モル)とクロロスルホニルイソシアナート14.2g(0.1モル)の混合溶液およびクロロシアン30.9g(0.5モル)とクロロスルホニルイソシアナート14.2g(0.1モル)の混合溶液を15分間かけて等モルずつ同時に滴下した。その後さらに反応液の還流温度(104℃～108℃)で9.5時間攪拌した。反応完結後におけるクロロスルホニルイソシアナート収率は90%(使用した三酸化硫黄基準。反応溶媒および原料希釈用として用いたクロロスルホニルイソシアナートは差し引いている。)であった。その後、常圧下において単蒸留を行ない、沸点106～108℃/1.013kPaの留分として、クロロスルホニルイソシアナート(純度99%)を94.1g得た。

(実施例2)

凝縮器を取り付けた容量100mlの四つ口フラスコにクロロスルホニルイソシアナート14.3g(0.1モル)を量り取り、攪拌しながら還流温度まで昇温した。この液中に、還流下、三酸化硫黄40.6g(0.5モル)とクロロスルホニルイソシアナート14.3g(0.1モル)の混合溶液およびクロロシア

ン 31.4 g (0.5 モル) とクロロスルホニルイソシアナート 28.4 g (0.2 モル) の混合溶液を 20 分間かけて等モルずつ同時に滴下した。その後さらに反応液の還流温度 (103℃～108℃) で 9 時間攪拌した。反応完結後におけるクロロスルホニルイソシアナート収率は 89% (使用した三酸化硫黄基準。反応溶媒および原料希釈用として用いたクロロスルホニルイソシアナートは差し引いている。) であった。その後、常圧下において単蒸留を行ない、沸点 106～108℃/1.013 kPa の留分として、クロロスルホニルイソシアナート (純度 99%) を 108.5 g 得た。

(比較例)

凝縮器を取り付けた容量 100 ml の四つ口フラスコにクロロスルホニルイソシアナート 14.3 g (0.1 モル) を量り取り、攪拌しながら還流温度まで昇温した。この液中に、還流下、三酸化硫黄 40.2 g (0.5 モル) およびクロロシアン 30.8 g (0.5 モル) を 60 分間かけて同時に滴下した。その際、クロロシアンは液状のものを一度気化させて流量計を通し、別の凝縮器で再び液化して反応系に滴下したが、クロロシアンの流量は一定しなかった。その後さらに反応液の還流温度 (95℃→109℃へ徐々に上昇) において 9 時間攪拌した。この時点におけるクロロスルホニルイソシアナート収率は 67% (使用した三酸化硫黄基準。反応溶媒および原料希釈用として用いたクロロスルホニルイソシアナートは差し引いている。) であった。

産業上の利用可能性：

本発明の製造方法によれば、省設備化を達成しつつ、温度制御の手間を省き、三酸化硫黄とクロロシアンから、操作性よく、高収率で、高純度のクロロスルホニルイソシアナートを製造することができる。

Exhibit B

DECLARATION

I, Sakiko SUGIHARA of c/o SHIGA INTERNATIONAL PATENT OFFICE,
GranTokyo South Tower, 1-9-2 Marunouchi, Chiyoda-ku, Tokyo 100-6620 Japan,
understand both English and Japanese, am the translator of the English document
attached, and do hereby declare and state that the amendments to the Specification
submitted in this Response to the Non-Final Office Action dated February 21, 2008,
correct translation errors made without intent to deceive in preparing a complete and
faithful English translation of International Patent Application No. PCT/JP04/019132
and that all statements made herein are true to the best of my knowledge.

Declared in Tokyo, Japan

This

Sakiko Sugihara

Sakiko SUGIHARA

May 16, 2008

Date

Exhibit C

[Claims]

[Claim 1]

A method for producing chlorosulfonyl isocyanate by reaction of sulfur trioxide with cyanogen chloride, wherein

chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate is used as a reaction solvent; and

sulfur trioxide and cyanogen chloride which are respectively diluted with the chlorosulfonyl isocyanate or the solution including chlorosulfonyl isocyanate are added at the same time to a reaction system in an almost equimolar amount under reflux.

[Claim 2]

A method for producing chlorosulfonyl isocyanate according to claim 1, wherein a reflux temperature is from 50 to 110°C.

[Document Type] Specification

[Title of Invention]METHOD FOR PRODUCING CHLOROSULFONYL ISOCYANATE

[Technical Field of the Invention]

[0001]

The present invention relates to a method for producing chlorosulfonyl isocyanate. More specifically, the present invention relates to a method for producing chlorosulfonyl isocyanate from sulfur trioxide and cyanogen chloride, in which the yield of the chlorosulfonyl isocyanate is high and the method has excellent operability.

[Background Art]

[0002]

Chlorosulfonyl isocyanate is industrially useful as an intermediate for producing pharmaceutical and agrochemical compounds, etc.

Conventionally, it is known that chlorosulfonyl isocyanate can be produced by the reaction of sulfur trioxide with cyanogen chloride, and several production methods thereof have been reported. For example, (a) Non-patent document 1 and Patent document 1 disclose a method in which sulfur trioxide is added to cyanogen chloride and reacted at a temperature of -50°C or lower. Also, (b) Patent document 2 and Patent document 3 disclose a method for reacting sulfur trioxide with cyanogen chloride at 100 to 200°C.

[0003]

However, the aforementioned method (a) is not preferable in view of cost, because it requires a large amount (from 1.5 to 3 times by mol) of cyanogen chloride to sulfur trioxide. Also, the method (a) is not preferable in view of safety, because it uses an excess of cyanogen chloride, which is toxic. Moreover, there are problems in that the yield of isolated chlorosulfonyl isocyanate is low at 60 to 62 %, and the quality, especially the purity, does not meet commercial requirements. Furthermore, when the aforementioned method (b) is used, it is not easy to control the flow rate of sulfur trioxide and cyanogen chloride added to a reaction system, and the yield of the obtained chlorosulfonyl isocyanate is low and the quality thereof is poor, similar to when the method (a) is used.

[0004]

In order to solve the aforementioned problems, (c) a method in which cyanogen chloride is added to sulfur trioxide and reacted while keeping the temperature of the reaction system at 20 to 50°C (Patent document 4), (d) a method in which sulfur trioxide and cyanogen chloride are added to a reaction system at the same time while keeping the

temperature of the reaction system at 10 to 50°C (Patent document 5), (e) a method in which sulfur trioxide and cyanogen chloride are reacted in a chlorinated hydrocarbon solvent (Patent document 6), and (f) a method in which cyanogen chloride is added to a mixture of sulfur trioxide and chlorosulfonyl isocyanate and reacted while keeping the temperature of the reaction system at -10 to 17°C (Patent document 7), and so forth have been proposed.

[0005]

More recently, (g) a method has been proposed, in which a residue in a tank is decomposed after separating chlorosulfonyl isocyanate by distillation and, at the same time, a low-boiling-point fraction obtained by separating a chlorosulfonyl isocyanate by distillation or by decomposing/distilling the residue in the tank is recovered, and the recovered liquid is reused by adding when reacting sulfur trioxide and cyanogen chloride (Patent document 8).

In those methods (c) to (g), chlorosulfonyl isocyanate having a comparably high yield (74 to 91%) and comparably high purity (90 to 98%) can be obtained by a comparably easy method.

[0006]

However, in the methods (c) to (f), because a cooling operation is needed to maintain the temperature within a suitable temperature range in which the temperature may be raised due to an exothermic reaction with sulfur trioxide and cyanogen chloride and the cost for the cooling equipment is high, these methods are not satisfactory as an industrial method. Also, in order to improve the yield in those methods, it is essential to obtain chlorosulfonyl isocyanate by thermo-decomposing chloropyrosulfonyl isocyanate as a by-product. Because an operation in which chloropyrosulfonyl isocyanate is thermo-decomposed is dangerous, complex, and costly, these methods have industrial problems.

Moreover, in the method (f), the number of processes is large, and a complex after-treatment is needed for salvaging and reusing the low-boiling-point fraction, and thus the method (f) has a problem in that a lot of equipment is required.

[0007]

[non-patent document 1] Chem. Ber., 89, 1071 (1956)

[patent document 1] West German Patent No. 928896

[patent document 2] European Patent No. 294613

[patent document 3] Swiss Patent No. 680292A5

[patent document 4] Japanese Unexamined Patent Application, First Publication
No. S 63-77855

[patent document 5] Japanese Unexamined Patent Application, First Publication
No. H1-228955

[patent document 6] Japanese Unexamined Patent Application, First Publication
No. H4-164063

[patent document 7] Japanese Unexamined Patent Application, First Publication
No. 2000-53630

[patent document 8] Japanese Unexamined Patent Application, First Publication
No. 2003-40854

[Disclosure of the Invention]

[Problems to be Solved by the Invention]

[0008]

In consideration of these circumstances surrounding the prior art, the object of the present invention is to provide a method for producing chlorosulfonyl isocyanate by a simple and easy operation, in which chlorosulfonyl isocyanate of high yield and high purity is obtained and the number of pieces of equipment is reduced.

[Means for Solving the Problem]

[0009]

As a result of extensive studies to solve the aforementioned problems, the inventors of the present invention found that it becomes easy to control the flow rate of sulfur trioxide and cyanogen chloride added to a reaction system by diluting sulfur trioxide and cyanogen chloride with chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate. Also, the inventors of the present invention found that the reaction is completed at the same time as by-products are thermo-decomposed under reflux of solvent by using chlorosulfonyl isocyanate or a solution containing chlorosulfonyl isocyanate as the reaction solvent, thereby leading to completion of the present invention.

[0010]

The present invention provides a method for producing chlorosulfonyl isocyanate by reaction of sulfur trioxide with cyanogen chloride, in which chlorosulfonyl isocyanate or a solution containing chlorosulfonyl isocyanate is used as the reaction solvent and sulfur trioxide and cyanogen chloride which are respectively diluted with chlorosulfonyl isocyanate or a solution containing chlorosulfonyl isocyanate are at the same time added to the reaction system under reflux in an almost equimolar amount.

A reflux temperature is preferably 50 to 110°C in the method for producing chlorosulfonyl isocyanate of the present invention.

[Effects of the Invention]

[0011]

By the production method of present invention, chlorosulfonyl isocyanate can be produced from sulfur trioxide and cyanogen chloride in which the yield and purity of the chlorosulfonyl isocyanate is high, the method has excellent operability, the amount of equipment is reduced, and time for controlling the temperature is saved.

[Embodiments of the Invention]

[0012]

The method for producing chlorosulfonyl isocyanate of the present invention is explained below in more detail.

The production method of the present invention is a method for producing chlorosulfonyl isocyanate by reaction of sulfur trioxide with cyanogen chloride.

[0013]

(1) Sulfur trioxide

Although the kind of sulfur trioxide used in the present invention is not especially limited provided that it is liquid, it is preferably a γ form because of high reactivity. Although the purity of sulfur trioxide used in the present invention is not especially limited, it is usually 90% by weight or more and preferable to 95% by weight or more. Also, the sulfur trioxide of the present invention may include a common polymerization inhibitor such as organosilicon, carbon tetrachloride, dimethyl sulfate, a boron compound, a phosphorus compound, and aromatic sulfonic acid. Although the content of the polymerization inhibitor is not especially limited, the content usually has a range within 0.001 to 1% by weight.

[0014]

(2) Cyanogen chloride

In the production method of the present invention, cyanogen chloride in the liquid state is used. Cyanogen chloride is industrially produced from hydrocyanic acid and chlorine, and is preferably dehydrated by dehydration or distillation. Also, although the purity of cyanogen chloride is not especially limited, the purity is usually 90% by weight or more and preferably 95% by weight or more.

[0015]

Usage of cyanogen chloride (in the case of a continuous system, usage by the hour) is usually 0.8 to 1.2 molar equivalents, preferably to 0.9 to 1.1 molar equivalents, to sulfur trioxide. Usage of cyanogen chloride which is less than 0.8 molar equivalents or more than 1.2 molar equivalents is not preferable, because production of by-products such as chloropyrosulfonyl isocyanate or 2, 6 - dichloro-1, 4, 3, 5-oxathiadiazine-4, 4-dioxide increase producing detrimental effects such as a reduction of yield or prolongation of the reaction completion time.

[0016]

(3) Method for producing chlorosulfonyl isocyanate

In the production method of the present invention, chlorosulfonyl isocyanate or a solution containing chlorosulfonyl isocyanate is used as a reaction solvent and sulfur trioxide and cyanogen chloride which are respectively diluted with chlorosulfonyl isocyanate or solution including chlorosulfonyl isocyanate are at the same time added to the reaction system under reflux by an almost equimolar amount.

[0017]

The purity of chlorosulfonyl isocyanate used as the reaction solvent and diluted solution of sulfur trioxide and cyanogen chloride is not especially limited. Chlorosulfonyl isocyanate having a high purity or a solution including chlorosulfonyl isocyanate may be used.

[0018]

Examples of a solution including chlorosulfonyl isocyanate include, but are not limited to, a reaction liquid which is used when producing chlorosulfonyl isocyanate and a starting fraction, main fraction, and residue from a tank which are distilled when purifying by distillation. Also, a part of the reaction liquid may be left in the reaction tank as solvent for a following batch.

[0019]

In the present invention, usage of chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate used as the reaction solvent is not especially limited provided that it is possible to control stirring and temperature. It is usually 0.2 to 1 molar equivalents, preferably to 0.2 to 0.5 molar equivalents, to the usage of sulfur trioxide.

[0020]

In the present invention, sulfur trioxide and cyanogen chloride are diluted with chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate.

By using sulfur trioxide and cyanogen chloride which are diluted with chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate, the diluted solutions of cyanogen chloride and sulfur trioxide are added under reflux while controlling the flow rate. Therefore, the production of by-products can be prevented and chlorosulfonyl isocyanate can be selectively obtained.

[0021]

Usage of sulfur trioxide and cyanogen chloride diluted with chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate is not especially limited when sulfur trioxide is diluted. When cyanogen chloride is diluted, a sufficient usage amount is that which does not cause cyanogen chloride to volatilize from chlorosulfonyl isocyanate as diluted solution. It is usually 0.2 to 1 molar equivalents, preferably to 0.2 to 0.5 molar equivalents, to the usage of cyanogen chloride.

[0022]

A method for adding sulfur trioxide diluted with chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate and cyanogen chloride diluted with chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate (hereinafter, they may be referred to as “raw material”) include a batch system and continuous system. In the case of a continuous system, a tank for completing the reaction may be provided in addition to a reaction tank.

[0023]

In the production method of the present invention, sulfur trioxide and cyanogen chloride which are respectively diluted with chlorosulfonyl isocyanate or solution containing chlorosulfonyl isocyanate are added at the same time to the reaction system under reflux in an almost equimolar amount. The term in “by an almost equimolar amount” in the description means that the charge ratio of sulfur trioxide to cyanogen chloride is from 0.8 : 1.2 to 1.2 : 0.8, preferably to 1.1 : 0.9 to 0.9 : 1.1, by molar ratio. The charge ratio means an amount per hour, in the case of a continuous system.

When a balance of the charge amount of the raw material is lost, the production of by-products such as chloropyrosulfonyl isocyanate or 2, 6 - dichloro-1, 4, 3, 5-oxathiadiazine-4, 4-dioxide increases producing detrimental effects such as a reduction of yield or prolongation of the reaction completion time.

[0024]

A charge temperature and a reaction temperature of the raw material indicate a reflux temperature of chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate as the solvent.

The reflux temperature is usually 50 to 110°C. In consideration of the decomposition of chloropyrosulfonyl isocyanate or 2, 6 - dichloro-1, 4, 3, 5-oxathiadiazine-4, 4-dioxide etc. for a short time, the reflux temperature is preferable to 100 to 110°C. In the case of using chlorosulfonyl isocyanate as the reaction solvent within the above reflux temperature range, cooling equipment is not needed and by-products including chloropyrosulfonyl isocyanate can be thermo-decomposed at the same time.

[0025]

A charge time for the raw material of the present invention is not especially limited provided that the condenser is allowed complete the reaction. In the case of using a batch system, the charge time is usually 0.25 to 2.5 hours. In the case of using a continuous system, the reaction may be performed by charging the amount of raw material according to the treatment capability of the equipment.

[0026]

In the production method of the present invention, it is preferable to provide a sufficient reaction time in order to produce chlorosulfonyl isocyanate having an excellent yield as the objective product and decompose by-products completely. The reaction time for the raw material to be sufficiently consumed and by-products to be decomposed is usually 0.5 to 15 hours, preferably 5 to 10 hours.

[0027]

(4) Operation of after-treatment

After the reaction, chlorosulfonyl isocyanate as the objective product can be isolated by refinement of the reaction liquid. The refinement of the reaction liquid after completing the reaction is usually performed using a normal distillation column under normal pressures or reduced pressure. Distillation conditions such as pressure, temperature, number of stages, or number of times are not especially limited. In the case of a continuous system, the reaction liquid is continuously ejected from the tank to complete the reaction and continuously distilled, and thus chlorosulfonyl isocyanate as the objective product can be isolated.

[Examples]

[0028]

The present invention will be explained below in more detail by reference to the following Examples and Comparative Example, but the invention should not be construed as being limited thereto.

[0029]

(Example 1)

14.3 g (0.1 mol) of chlorosulfonyl isocyanate was placed in a 100 ml-four-neck flask with a condenser and the temperature thereof was raised to the reflux temperature while stirring. To this solution, a mixed solution of 40.4 g (0.5 mol) of sulfur trioxide and 14.2 g (0.1 mol) of chlorosulfonyl isocyanate and a mixed solution of 30.9 g (0.5 mol) of cyanogen chloride and 14.2 g (0.1 mol) of chlorosulfonyl isocyanate were added dropwise at the same time in the same molar equivalents for 15 minutes under reflux to obtain a reaction liquid. Then, the reaction liquid was stirred for 9.5 hours at the reflux temperature (from 104 to 108°C). The yield of chlorosulfonyl isocyanate was 90% after completing the reaction, in which the used sulfur trioxide was based on and the amount of chlorosulfonyl isocyanate used as reaction solvent and solution for diluting the raw material was removed. Then, simple distillation was performed under normal pressures to obtain 94.1 g (the purity of 99 %) of chlorosulfonyl isocyanate as the fraction by boiling at 106 to 108°C under 1.013 kPa.

[0030]

(Example 2)

14.3 g (0.1 mol) of chlorosulfonyl isocyanate was placed in a 100 ml-four-neck flask with a condenser and the temperature thereof was raised to the reflux temperature while stirring. To this solution, a mixed solution of 40.6 g (0.5 mol) of sulfur trioxide and 14.3 g (0.1 mol) of chlorosulfonyl isocyanate and a mixed solution of 31.4 g (0.5 mol) of cyanogen chloride and 28.4 g (0.2 mol) of chlorosulfonyl isocyanate were added dropwise at the same time in the same molar equivalents for 20 minutes under reflux to obtain a reaction liquid. Then, the reaction liquid was stirred for 9 hours at the reflux temperature (from 103 to 108°C). The yield of chlorosulfonyl isocyanate was 89% after completing the reaction, in which the used sulfur trioxide was based on and the amount of chlorosulfonyl isocyanate used as reaction solvent and solution for diluting the raw material was removed. Then, simple distillation was performed under normal pressures to obtain 108.5 g (the purity of 99 %) of chlorosulfonyl isocyanate as the fraction by boiling at 106 to 108°C under 1.013 kPa.

[0031]

(Comparative Example)

14.3 g (0.1 mol) of chlorosulfonyl isocyanate was placed in a 100ml-four-neck flask with a condenser and the temperature thereof was raised to the reflux temperature while stirring. To this solution, a mixed solution of 40.2 g (0.5 mol) of sulfur trioxide and 30.8 g (0.5 mol) of cyanogen chloride were added dropwise at the same time in the same molar equivalents for 60 minutes under reflux to obtain a reaction liquid. In this operation, cyanogen chloride in the liquid was vaporized, passed through a flow meter, and added dropwise to the reaction system by another condenser, but the flow rate of cyanogen chloride was not fixed. Then, the reaction liquid was stirred for 9 hours at the reflux temperature of the reaction liquid which was gradually raised from 95 to 109°C. At this point, the yield of chlorosulfonyl isocyanate was 67%, in which the used sulfur trioxide was based on and the amount of chlorosulfonyl isocyanate used as reaction solvent and solution for diluting the raw material was removed.

[Document Type] Abstract

[Abstract]

[Problem to be Solved by the Invention]

A method for producing chlorosulfonyl isocyanate, in which the yield and purity of the chlorosulfonyl isocyanate is high, the method has excellent operability, and number of equipments is reduced, is provided.

[Means for Solving the Problem]

A method for producing chlorosulfonyl isocyanate by reaction of sulfur trioxide with cyanogen chloride, wherein chlorosulfonyl isocyanate or a solution including chlorosulfonyl isocyanate is used as a reaction solvent, sulfur trioxide and cyanogen chloride which are respectively diluted with the chlorosulfonyl isocyanate or the solution including chlorosulfonyl isocyanate are added at the same time to a reaction system in an almost equimolar amount under reflux.

[Selected Drawing] No Drawing

Exhibit D

DECLARATION

I, Sakiko SUGIHARA of c/o SHIGA INTERNATIONAL PATENT OFFICE,
GranTokyo South Tower, 1-9-2 Marunouchi, Chiyoda-ku, Tokyo 100-6620 Japan,
understand both English and Japanese, am the translator of the English document
attached, and do hereby declare and state that the attached English document contains
an accurate translation of the official certified copy of Japanese Patent Application
No(s). 2003-417611 and that all statements made herein are true to the best of my
knowledge.

Declared in Tokyo, Japan

This

Sakiko Sugihara

Sakiko SUGIHARA

May 16, 2008

Date

Exhibit E

[0011]

(First Process) The first process includes a process in which a cyanogen chloride (material A) and sulfuric anhydride (material B) are mixed and condensed to obtain a condensation reaction solution including chlorosulfonyl isocyanate. The object of this process is that a cyanogen chloride and sulfuric anhydride are reacted and converted to chlorosulfonyl isocyanate with good selectivity; and that the concentration of chlorosulfonyl isocyanate in the reaction solution is at least 80% by mass or more and preferable 90% by mass or more after the reaction. In order to achieve this object, it is preferably to lower the production rate of chloropyrosulfonyl isocyanate or 2,6-dichloro-1,4,3,5-oxathiadiazine-4,4-dioxide as the main by-product. To do that, as described later, the condensation reaction temperature or the reaction rate of a cyanogen chloride and sulfuric anhydride may be controlled.

Exhibit F

DECLARATION

I, Sakiko SUGIHARA of c/o SHIGA INTERNATIONAL PATENT OFFICE,
GranTokyo South Tower, 1-9-2 Marunouchi, Chiyoda-ku, Tokyo 100-6620 Japan,
understand both English and Japanese, am the translator of the English document
attached, and do hereby declare and state that the attached English document contains
an accurate translation of the official certified copy of paragraph [0011] in Japanese
Patent Publication No(s). 2003-040854 and that all statements made herein are true to
the best of my knowledge.

Declared in Tokyo, Japan

This

Sakiko Sugihara

Sakiko SUGIHARA

May 16, 2008

Date